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A method for analyzing the non-stationary nucleation and overall transition kinetics: A case of water

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Abstract

We present the statistical method as a direct extension of the mean first-passage time concept to the analysis of molecular dynamics simulation data of a phase transformation. According to the method, the mean first-passage time trajectories for the first (i 1) as well as for the subsequent (i 2, 3, 4,) nucleation events should be extracted that allows one to calculate the time-dependent nucleation rate, the critical value of the order parameter (the critical size), the waiting times for the nucleation events, and the growth law of the nuclei - i.e., all the terms, which are usually necessary to characterize the overall transition kinetics. There are no restrictions in the application of the method by the specific thermodynamic regions; and the nucleation rate parameters are extracted according to their basic definitions. The method differs from the Wedekind-Bartell scheme and its modification [A. V. Mokshin and B. N. Galimzyanov, J. Phys. Chem. B 116, 11959 (2012)], where the passage-times for the first (largest) nucleus are evaluated only and where the average waiting time for the first nucleation event is accessible instead of the true steady-state nucleation time scale. We demonstrate an efficiency of the method by its application to the analysis of the vapor-to-liquid transition kinetics in water at the different temperatures. The nucleation rate/time characteristics and the droplet growth parameters are computed on the basis of the coarse-grained molecular dynamics simulation data.

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